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Aqueous chemistry of anthropogenically contaminated Bengaluru lakes



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Abstract

Anthropogenic activities impact the major ion composition of fresh water sources. The inorganic species are likely to be influenced by mineral dissolution, mineral precipitation, redox and ion-exchange reactions in the contaminated lakes. Owing to paucity of research, this study examines the influence of bio-geochemical reactions on the major ion composition of sewage contaminated Bengaluru lakes. The selected lakes represent water bodies in the major valley systems of the city that are polluted by partly treated sewage and stormwater runoffs. Hydrogeochemical facies of the lake samples showed that enrichment of Mg^{2+} , Na^+ , Cl^- and SO_4^{2-} ions from anthropogenic contamination altered the chemical type of the lake-water. Examination of processes influencing the major ion composition of surface waters indicated that evaporation than rock-weathering tends to influence the chemical composition of the Bengaluru lakes. Precipitation of carbonate minerals in the alkaline pH contributed to the deficiency of alkaline earth ions, while dissolution of anthropogenic gypsum enhanced the SO_4^{2-} ion concentration of the lakes. Solute diffusion from lake water into the pore solutions of sediments and ion-exchange reactions between monovalent ions of sediments and divalent cations in lake water are additional pathways that influence the major ion composition of the contaminated lakes. Besides alterations in major ion composition, organic contamination, biochemical reactions associated with photosynthesis activity of algae and release of toxic Al³⁺ ions from mineral dissolution are consequences of the lake contamination.

Keywords: Anthropogenic, Lakes, Geochemistry, Pollution, Wastewater

Introduction

The natural factors of lithology, weathering of drainage catchments, precipitation, climate and distance to marine environment influence the major ion composition of freshwater resources [1-4]. The geochemical processes of dissolution/precipitation, ion-exchange, evaporation/ crystallization and redox transformations control the fate of the inorganic species in the fresh water sources [4-8].

Anthropogenic contamination also impacts the major ion composition of freshwater resources. Contamination by fertilizers, animal waste, municipal and industrial sewage have altered the major ion composition (SO_4^{2-} , NO_3^- , Cl^- , and K^+) of the river waters in Guizhou Province, China [7]. Use of manure and chemical fertilizers have contaminated the surface reservoirs of northwest Iran with NO_3^- , CI^- , HCO_3^- and Na^+ ions [8]. Anthropogenic activities related to agriculture have led to strong correlations between TDS (total dissolved solids) and major ion ratios [7, 9]. Agricultural fertilizers caused significant increase in Na^+ , SO_4^{2-} , K^+ , CI^- , NO_3^- , and Mg^{2+} ion concentrations and altered the chemical ratios in the Seine river basin, France [2]. Nitrate pollution from agriculture and floating gardens led to a reverse correlation between nitrate and other major ions in the Dal lake samples of Gagribal and Nagin basins, India [10]. Dissolution of anthropogenic gypsum arising from idols immersed in urban lakes contributed to the Ca^{2+} ion concentrations of such water bodies [11].

Black water is rich in major cations and anions [12-15]; sewage contamination would thus alter the major ion composition of the contaminated lakes and rivers. Base flow solute concentrations (SO₄², Cl⁻,



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 HCO_3^- , and Na⁺) were observed to increase with the degree of urbanization in the Atlanta Metropolitan Region, USA, suggesting that septic tank systems and leaky sewer line functioned as pervasive low-level non-point sources of contamination affecting the chemistry of shallow groundwater. A strong and unusual basin-wide correlation between Na-K-Cl within the river basin reflected the widespread input of electrolytes present in human wastes and wastewater [16]. of Bagmati river water The composition in Kathmandu, Nepal, suggested that sewage effluent entering the river had a major effect on its major ion composition. All the chemical parameters showed strong correlations with population density suggesting that population density is the most likely driver of chemical load in drainage waters of the river [17]. Also, the microbial degradation of organic C and oxidation of NH_4^+ ions [12, 18, 19] will contribute to HCO₃⁻ and NO₃⁻ concentrations of sewage contaminated lakes and rivers. Occurrence of alkaline pH from photosynthetic metabolism of algae [20-23] can induce carbonate mineral precipitation [24, 25] and impact the alkaline earth (Ca^{2+} , Mg^{2+}) and HCO_3^{-} ion composition of sewage contaminated water bodies.

Earlier studies have highlighted the influence of anthropogenic contamination on the major ion composition of freshwater resources. Subsequent to their deposition, the inorganic species will possibly be influenced by dissolution/precipitation, ion-exchange, evaporation/crystallization and redox transformations in an environment affected by microbial and algal activities. The influence of bio-geochemical reactions on the major ion composition of contaminated freshwater resources has not been studied. The present study hence examines the influence of bio-geochemical reactions on the major ion composition of sewage contaminated Bengaluru lakes. The selected lakes represent water bodies in the major valley systems of Bengaluru city that are contaminated by partly treated sewage and stormwater runoffs.

Study area

Bengaluru is located in southeastern part of Karnataka extending from north latitude $12^{\circ}39'$ 32'' to $13^{\circ}14'13''$ and East longitude $77^{\circ}19'44''$ to $77^{\circ}50'13''$ at an altitude of 920 m above mean sea level. Most of Bengaluru lakes are irrigation tanks built in sixteenth century and are hydrologically connected to form a chain of lakes in a given valley system. Rapid urbanization of the city from 1980s has diminished the number of lakes; further insufficient recharge, inflow of partially treated sewage, stormwater runoffs and effluents have contaminated the existing lakes [26–28].

The lake beds are composed of residually derived red sandy soil that are overlaid by silt deposits of varying thickness [29, 30]. The silt fraction of red soils deposited in lakes from rainwater runoffs are expected to be composed of kaolinite and silica minerals [29]. The area is mainly drained by part of Arkavathi river catchment to the west and south Pennar river to the east. The two main catchments are further sub-divided into three major valley watersheds namely the Hebbal Valley, Vrushabhavathi Valley, and Koramangala and Challaghatta (KC) Valley. Each of these valleys are endowed with two lake series, and each lake series in turn comprise of many lakes. The flow of water in each lake series is along a network of drains which facilitates surplus water from the lakes to flow into the downstream lake. Drain is a naturally or man-made channel which connects the lentic water bodies for the purpose of conveying the surplus water on land, particularly rainfall runoff. In Bengaluru City, most of the nonrejuvenated drains however carry sewage to water bodies and are chocked by siltation and weeds. The catchment areas of Hebbal valley, Vrushabhavathi valley and KC valley correspond to 207, 165, and 255 km^2 respectively [31].

Among the lakes examined in this study, Puttenahalli lake (PU; KC valley) is the end member of the Puttenahalli lake series that does not drain further. Ulsoor lake (UL; KC Valley) is part of the Varthur lake series and the end-member of this series (Varthur lake) drains into the South Pinakini river. Sankey lake (SK; Vrushabhavathi Valley) is standalone lake and is fed by rainwater runoffs and stormwater drains. Nagawara lake (NG; Hebbal Valley) is part of the Yellamallappa Chetty lake series; the end member (Kacharakanahalli lake) of this lake series does not drain further.

The areal extent, elevation and coordinates (coordinates are obtained using Google Earth Pro software) of the lakes are presented in Table 1. The location of the four lakes and primary storm-water drains in the major valley systems of Bengaluru city are illustrated in Fig. 1. The maximum and average depths of PU, UL and NG lakes were measured in April-May 2019. The depth of each lake was measured at multiple points (4 to 9) with chain and solid metal ball (4 kg) assembly in consultation with local experts having knowledge of the lake topography. The maximum depths of PU, UL and NG lakes correspond to 2.1, 3.3, and 2.0 m respectively, while, their average depths are 1.43, 2.5 and 1.41 m respectively. The maximum and average depth of SK lake corresponds to 9.0 m [33] and 2.7 m [32] respectively.

Name of the lake	Areal extent (ha) ^a	Elevation (m) ^a	Location ^b
Sankey lake	14.99	917	Lies between latitudinal parallels of 13°0'27" N and 13°0'44" N and longitudinal meridians of 77°34'22" E and 77°34'37" E
Nagawara lake	23.49	896	Lies between latitudinal parallels of 13°2'47" N and 13°2'35" N and longitudinal meridians of 77°36'57" E and 77°36'43" E
Puttenahalli lake	5.37	915	Lies between latitudinal parallels of 12°53'29" N and 12°53'21" N and longitudinal meridians of 77°35'9" E and 77°35'17" E
Ulsoor Lake	40.59	905	Lies between latitudinal parallels of 12°58'38" N and 12°59'01" N and longitudinal meridians of 77°36'57" E and 77°37'21" E

Table 1 Areal extent, elevation and coordinates of the lakes

^a [32]

^b Google Earth Pro software

Field collection of lake water and wastewater samples

Collection of water column and sediment samples

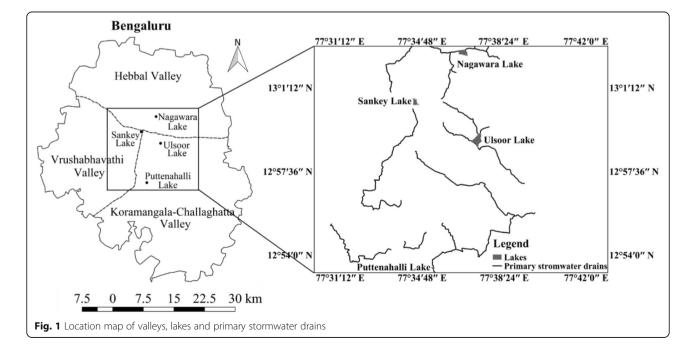
The water column profile of UL, PU and NG lakes was established by collecting samples at varying depths (0.5 to 2.5 m, Fig. 2) during April-May 2019. The DO (dissolved oxygen measured using Eutech instruments, Highland Park, New Jersey, USA) and temperature probes were attached to the chain and ball assembly that facilitated in-situ measurements at desired depths. Water samples were collected to a maximum depth of 2.5 m (lake bed depth = 3.5 m from surface) at UL Lake, from 1.0 m depth (lake bed depth = 2.0 m) at NG Lake and from 1.5 m depth (lake bed depth = 2.5 m) at PU Lake. The water column profiles of UL, PU and NG lakes (Fig. 2) show that temperature, pH, DO, TDS and HCO₃⁻ values of a given lake vary insignificantly with depth indicative of a well-mixed system.

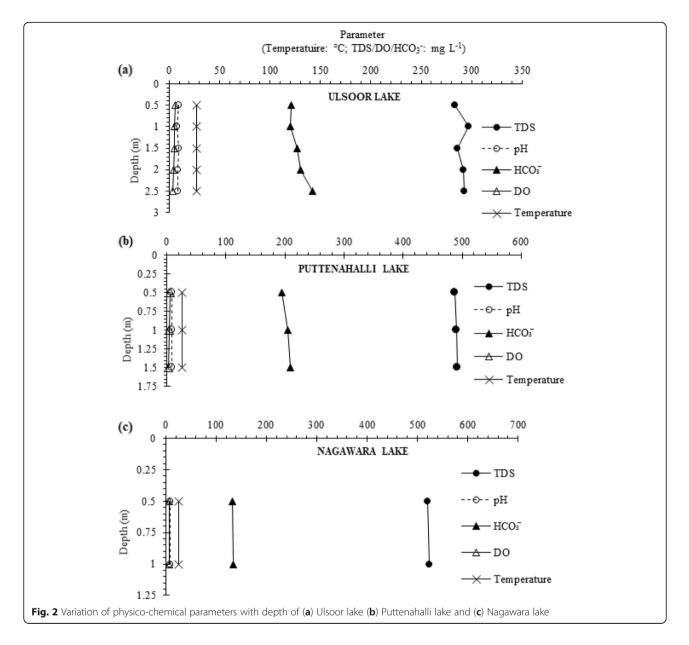
Sediments were collected at each lake (Figs. 3a to c) using Van Veen Grab sampler. The coordinates of the sampling points were recorded using a handheld GPS device (Garmin Etrex 30). The sediment samples were stored in labelled zip-lock covers. Both water and sediment samples were shipped at 4 °C and preserved at 4 °C in the laboratory.

Collection of discharge samples

PU lake has 7 discharge points (Fig. 3a); of these, PUIN1 is the outfall of a sewage treatment plant of a residential apartment complex [34]. The other 6 discharge points (PUIN2 to PUIN7) are outlets of stormwater drains. Stormwater outlets also discharged into UL lake (ULIN1 and ULIN2, Fig. 3b), NG lake (NGIN1 and NGIN2, Fig. 3c) and SK lake (SKIN1 to SKIN5, Fig. 3d).

As only PUIN1 and ULIN1 were active during the study period, samples were collected from these discharge points. Two samples were collected from PUIN1





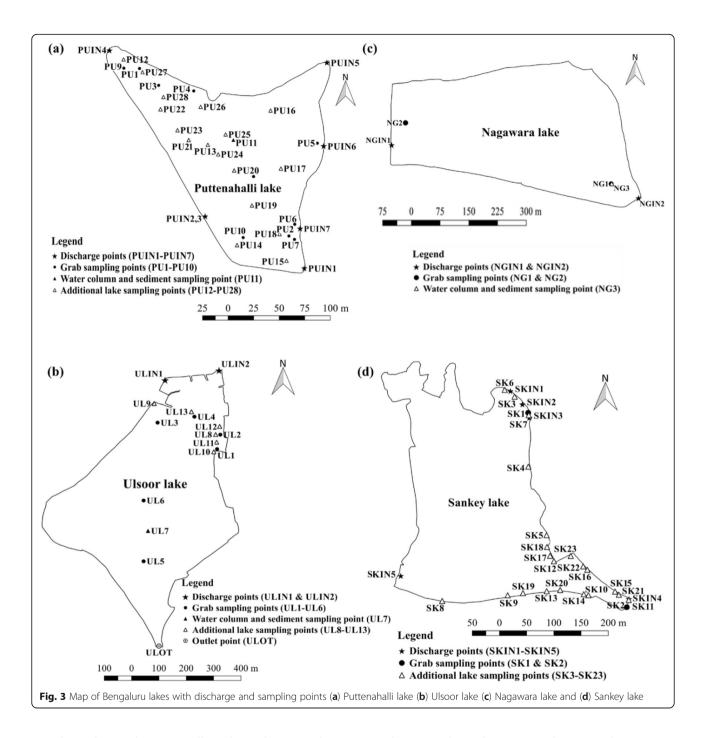
(March and June 2018) with one sample collected from ULIN1 (June 2018). At each sampling location (PUIN1 and ULIN1), three, 1-L samples were collected for various analysis. Of the 3 samples, one sample was acidified with concentrated nitric acid for cation analysis, while, the two non-acidified samples were used for COD (Chemical Oxygen Demand), $\rm NH_4^+$, and anion measurements.

Collection of lake samples

Nineteen grab samples were collected from the four lakes (Fig. 3) during January to June 2018. The sampling was performed between 09:00 and 15:00 h at all the lakes. The selection of sampling locations in a lake was guided by the availability of boat facility. Availability of

boat facility for limited period restricted the number of samples that were collected from NG lake. Nonavailability of boat facility at SK lake confined the sampling to the lake periphery.

The sampling frequency of a given lake was guided by the occurrence of fish-kill event during the study period (January 2018 to June 2018); significant algal bloom is often associated with fish-kill. Samples were collected from PU lake in January (non-algal bloom condition) and March 2018 (fish kill was reported on 21 March 2018 [35]) at 10 locations (Fig. 3a). Likewise, samples were collected from UL lake in January 2018 and June 2018 (fish kill reported on 30 May 2018) at 6 locations (Fig. 3b). At SK and NG lakes, fish kill was not reported during the study period and hence the lakes were not re-



sample; grab samples were collected at 2 locations from NG lake (Fig. 3c) and at 2 locations from SK lake (Fig. 3d) in January 2018. Grab samples were collected at shallow depths (15–90 cm) from each lake using a stainless-steel bailer.

Five batches of water samples were collected at each sampling location of a given lake. Of the 5 batches, two samples were acidified with concentrated nitric acid for cation analysis, while, the three non-acidified samples were used for COD, $\rm NH_4^+$, and anion measurements. All

samples were shipped at $4\,^\circ\mathrm{C}$ and preserved at $4\,^\circ\mathrm{C}$ in the laboratory.

Besides sample collection during January 2018 to June 2018, additional sampling was performed at SK, PU and UL lakes, to monitor pH, DO and TDS parameters. Samples were collected from twenty-one additional locations at SK lake in July 2018, September 2018, February 2019, April 2019 and May 2019. Likewise, samples were collected from six additional locations at UL lake in July 2018 and February 2019. Samples were collected from

seventeen additional locations at PU lake in September 2018, April 2019 and May 2019. The additional sampling locations of PU lake (PU12 to PU28), UL lake (UL8 to UL13) and SK lake (SK3 to SK23) are included in Fig. 3a, b and d respectively. Both lake and discharge samples were collected on dry days of the sampling period. The statistical distribution of measured parameters (Table 2) includes the data from additional sampling and water column profile of the lakes.

Methods of analysis

Tests with grab and discharge samples

Electrical conductivity (EC), pH and DO contents of lake water and discharge (PUIN1 and ULIN1) samples were measured in the field using portable EC, pH, and DO meters. All other parameters were measured in the laboratory. The EC values are represented as TDS using an appropriate relation. The COD concentrations of the lake water and PUIN1 and ULIN1 samples were determined with unfiltered samples using silver sulfatesulfuric acid closed reflux method [38]. The other parameters were measured with filtered (0.45-µm filter, Merck Millipore, USA) samples. The concentrations of Mg^{2+} , Ca^{2+} , Na^+ and K^+ ions and heavy metal ions (Fe²⁺, Mn²⁺, Cu²⁺, Pb²⁺, Zn²⁺, Al³⁺ and Cd²⁺) in the acidified lake water and PUIN1 and ULIN1 samples were measured using atomic absorption spectrometer (Thermo AAS, ICE 3000, Thermo Fisher Scientific, Waltham, Massachusetts, USA).

The NH4⁺ and anion concentrations were measured with non-acidified (filtered) samples. The SO₄²⁻ ion concentration was measured [38] by turbidimetric method at 420 nm absorbance using UV-Visible spectrophotometer (Thermo Evolution 201, Thermo Fisher Scientific, Waltham, Massachusetts, USA). The NO₃⁻ and NH₄⁺ ion concentrations were determined by ionchromatograph (Dionex ICS 2000, Thermo Fisher Scientific, Waltham, Massachusetts, USA). The Cl- and HCO_3^{-}/CO_3^{2-} ion concentrations were determined using automatic titrator (Metrohm Titrino Plus 877, Metrohm AG, Herisau, Switzerland). The ionic ratio method helps to assess the role of mineral dissolution in influencing the ion composition of water samples. When weathering and dissolution of calcite, dolomite and gypsum significantly contribute to the chemical composition of water, the data points in $(Ca^{2+} + Mg^{2+})$ versus $(HCO_3^{-} + SO_4^{2-})$ chart will plot on/close to the 1:1 line [8]. Piper chart provides visualization of dominant cations and anions present in water samples. The chart reveals similarity and differences between water samples based on the hydrochemical facies. The z is function of lithology, solution kinetics and flow path of water [39].

Chlorophyll-a (Chl-a) concentrations in lake water samples were determined by the spectrophotometer method [38, 40] using 5 L samples. For Chl-a determination, appropriate aliquot of samples was concentrated by passing through Whatman GF/C filters. Prior to extraction, filters were homogenized to enhance extraction

 Table 2 A statistical summary of measured parameters of lake and MWW samples

Parameter	Mixed waste	water samples		Lake samples	5		Average	Average
	Minimum	Median	Maximum	Minimum	Median	Maximum	values (mg L ⁻¹) of Bengaluru city groundwater samples [36]	values (mg L ⁻¹) of IISc groundwater samples [37]
рН	7.15	7.18	7.9	6.0	8.8	10.3	-	_
HCO_{3}^{-} (mg L ⁻¹)	109	305	551	80	109	344	341	165
Cl^{-} (mg L^{-1})	95	100	102	36	66	102	181	50
SO_4^{2-} (mg L ⁻¹)	6	10	20	7	40	319	88	12
NO_{3}^{-} (mg L ⁻¹)	0.5	0.7	2.5	0.0	3.5	9.0	53.0	—
Ca^{2+} (mg L ⁻¹)	46	53	78	17	25	90	117	38
Mg^{2+} (mg L ⁻¹)	20	20	24	7	14	40	53	15
Na^{+} (mg L^{-1})	73	87	95	20	57	92	97	39
K^{+} (mg L^{-1})	17	19	20	8	13	16	6	-
$NH_4^+ (mg L^{-1})$	16	29	50	0.0	0.0	4	-	_
TDS (mg L^{-1})	591	683	811	173	359	882	770	234
COD (mg L^{-1})	54	83	99	12	42	67	-	-
DO (mg L^{-1})	0.1	0.4	0.5	4.1	7.5	15.0	-	-
Chl-a (μ g L ⁻¹)	-	-	-	BDL	4	19	-	-

BDL Below detection limit

efficiency. Extraction was performed using 90% acetone followed by centrifugation to produce a supernatant for spectrophotometric determination. The absorbance was recorded at four different wavelengths (630, 645, 665 and 750 nm) for measuring Chl-a concentration. The detection limit of the method is $0.02 \,\mu g \, L^{-1}$. Trophic state index (TSI) based on Chl-a concentration is calculated to assess lake's trophic state in the terms of algal productivity. Trophic state indicates anthropogenic influence on water quality and the ecological functioning of lakes. The scale of TSI ranges from 0 to 100. Each major division represents a doubling in algal biomass. The index number can be calculated from any of several parameters, including Secchi disk transparency, chlorophyll, and total phosphorus [41].

The COD concentrations of PUIN1 (treated sewage) and ULIN1 (stormwater runoff) samples (Table 2) range from 54 to 99 mg L⁻¹ (median = 83 mg L⁻¹). The DO concentrations range from 0.10 to 0.48 mg L⁻¹ (median = 0.42 mg L⁻¹) and NH₄⁺ ion concentrations range from 16 to 50 mg L⁻¹ (median = 29 mg L⁻¹). The anoxic nature of the samples hindered the oxidation of NH₄⁺ ions resulting in low concentrations of NO₃⁻ ions. Based on the COD, DO and NH₄⁺ concentrations, the stormwater runoffs and treated sewage discharging into the Bengaluru lakes are referred as mixed wastewater (MWW) in further discussions.

Tests with sediment samples

The gravimetric moisture content of the sediments was determined by oven-drying them at 105 °C for 24 h. The organic matter of the sediments was determined by Walkley Black method. The EC was determined by agitating the sediments with ultra-pure water (sediment: water ratio = 1:2.5) for 60 min. After centrifugation, the EC of supernatant was measured and converted to TDS values. Water leaching tests were performed with the lake sediments in accordance with ASTM procedure [42]. The water leachate was examined for Ca²⁺, Mg²⁺, Na⁺, K⁺, Cl⁻, HCO₃⁻ and SO₄⁻ ions by the previously described procedures.

The sediment residue from water leaching test was agitated again with ultra-pure water (200 mL) for 2 h to remove residual salts. The suspension was centrifuged, and the EC of the washings ranged between 25 and 45 μ S cm⁻¹. The exchangeable cation content of the moistwashed residue was determined using ammonium acetate extraction method [43]. The concentrations of displaced Ca²⁺, Mg²⁺, Na⁺, K⁺ ions in the ammonium acetate extracts were estimated using atomic absorption spectrometer. Corrections were applied for residual cation concentrations persisting in the moist-washed sediments.

Results and discussion

Chemical composition of lake water samples

Table 2 summarizes the statistical distribution of chemical parameters of the lake and MWW samples. The lake samples are characterized with median pH of 8.8. The median cation concentrations of the lake samples vary as: $Na^+ > Ca^{2+} > Mg^{2+} \approx K^+ > NH_4^+$. Likewise, the median anion concentrations vary as: $HCO_3^- > Cl^- > SO_4^{2-} >$ NO₃⁻. The lake samples showed measurable concentrations of Al^{3+} and Zn^{2+} ions, while, the strengths of other heavy metal ions (Fe²⁺, Mn²⁺, Cu²⁺, Pb²⁺ and Cd²⁺) were below the detection limit of the instrument (< 0.01 mg L^{-1}). The Al³⁺ ion concentrations in the lake samples ranged from 0.14 to 0.39 mg L^{-1} ; further, the Al³⁺ ion concentrations of twelve samples were greater than its permissible limit [44] in drinking water (0.2 mg L^{-1}) . The MWW samples did not show the presence of Al³⁺ ions. It is hence likely that dissolution of oxide coatings on particle surfaces and/or dissolution of mineral particle edges by humic acid are possibly responsible for presence of Al^{3+} ions in the lake samples [45]. The Zn^{2+} ion concentrations in the lake samples ranged from 0.011 to 0.081 mg L^{-1} and are below the permissible limit in drinking water $[15 \text{ mg L}^{-1}; 44]$. Comparatively, the Zn²⁺ ion concentrations in the MWW samples ranged between 0.032 to 0.035 mg L^{-1} .

Multiple correlation analysis

The lake water data was analysed for multiple correlations among the measured variables (Table 3). The strong positive correlation between DO and pH (R =0.74) is attributed to the production of DO and OH⁻ ions during intense photosynthesis activity. Likewise, the strong negative correlation (R = -0.72) between pH and HCO₃⁻ ions arises from bicarbonate consumption and release of OH⁻ ions during intense algal photosynthetic activity [19, 22]. The strong negative correlation between Ca^{2+} and Mg^{2+} ion concentrations with pH (R = -0.64to -0.62) apparently arise as the alkaline pH reduces the divalent cation concentrations from carbonate precipitation reactions (discussed in section 4.3.2). The strong positive correlation (R = 0.69) between pH and CO_3^{2-} ions is attributed to HCO_3^- to CO_3^{2-} transformation in the alkaline pH environment (pH \ge 8.35). The strong correlation between Ca²⁺ and HCO₃⁻ ions arises from their tendency to form calcium carbonate in an alkaline pH environment. The strong correlation between Ca²⁺ and SO₄²⁻ ions is attributed to dissolution of anthropogenic gypsum. The positive correlation between Chl-a and pH (R = 0.74) supports the hypothesis that algal photosynthesis renders the lake water alkaline. The positive correlation between Chl-a and DO (R = 0.59) is attributed to production of DO by photosynthetic algae. The negative correlation between Chl-a and HCO₃⁻ ions

Table 3 Multiple correlation analysis for grab samples

-	TDS	рН	CO3 ²⁻	HCO3-	CI ⁻	SO4 ²⁻	NO_3^-	Ca ²⁺	Mg ²⁺	Na ⁺	K+	DO	Chl-a
TDS	1												
рН	-0.06	1											
CO3 ²⁻	- 0.14	0.69	1										
HCO3 ⁻	0.49	-0.72	-0.57	1									
CI ⁻	0.25	0.25	0.68	0.12	1								
SO4 ²⁻	0.32	-0.59	-0.59	0.71	-0.05	1							
NO_3^-	0.45	-0.58	-0.79	0.64	-0.43	0.77	1						
Ca ²⁺	0.36	-0.64	-0.29	0.87	0.30	0.67	0.50	1					
Mg^{2+}	0.61	-0.62	-0.36	0.86	0.16	0.69	0.61	0.84	1				
Na ⁺	0.19	0.39	0.63	0.06	0.90	-0.04	-0.37	0.27	0.06	1			
K^+	-0.06	0.19	0.43	0.04	0.73	0.14	-0.36	0.29	-0.03	0.81	1		
DO	-0.12	0.74	0.73	-0.71	0.42	-0.66	-0.72	- 0.57	-0.68	0.47	0.33	1	
Chl-a	0.10	0.74	0.44	-0.51	0.27	-0.20	- 0.27	-0.42	- 0.47	0.36	0.30	0.59	1

is attributed to consumption of bicarbonate ions by algae.

Chl-a concentrations in the lake samples during nonalgal bloom condition ranged from < 0.02 to 0.022 μ g L⁻¹ (PU1 & PU2, SK1 & SK2; Table 3). Comparatively, Chl-a concentrations in the lake samples are significantly higher during algal bloom condition and ranged from 2.8 to 18.8 μ g L⁻¹ (PU3–PU10, NG2, UL1–UL6; Table 3). The TSI of the lakes is estimated from Chl-a values using the equation [41, 46],

$$TSI (Chl-a) = 9.81 \ln (Chl-a) + 30.6$$
 (1)

Values of TSI < 30 corresponds to oligotrophic state and values of TSI > 50 corresponds to eutrophic state [46]. The TSI values of majority (13 out of 19) of lake samples range between 41 to 59 (Table 4) indicative of near eutrophic or eutrophic state. The eutrophic status of the lakes supports the hypothesis that photosynthetic metabolism of algae is responsible for strong correlations between the lake parameters (Table 3).

Eutrophic state indicates substantial growth of algal population which can disrupt the DO regime of the lake. Substantial algal growth causes oxygen supersaturation during photosynthetic period (maximum DO concentration = 15 mg L^{-1} ; Table 2) and decrease in DO during night time because of algal respiration. Periods of oxygen supersaturation is detrimental and leads to fish mortality [47]; and hypoxia caused by algal bloom collapse is often associated with fish kill [48].

Anthropogenic influence on major ion composition of Bengaluru lakes

Table 2 includes the average composition of major ions in the groundwater of Bengaluru city [36] and Indian Institute of Science (IISc) campus, Bengaluru [37]. The major cation and anion concentrations of IISc ground-water samples are 0.36–0.53 and 0.29–0.61 times lower than those of the Bengaluru city groundwater samples and are 0.14 to 0.72 times lower than the median cation concentrations of MWW samples (Table 2). The major anion concentrations of IISc groundwater samples are

Table 4 Trophic state of Bengaluru lakes

Sample ID	Chl-a (μ g L ⁻¹)	TSI (Chl-a)	Trophic state ^a
PU1	< 0.02	0	Oligotrophic
PU2	0.022	0	Oligotrophic
PU3	9.7	53	Eutrophic
PU4	3.7	43	Near eutrophic
PU5	5.9	48	Near eutrophic
PU6	5.7	48	Near eutrophic
PU7	7.8	51	Eutrophic
PU8	10.9	54	Eutrophic
PU9	4.3	45	Near eutrophic
PU10	17.2	59	Eutrophic
UL1	5.0	46	Near eutrophic
UL2	_	_	_
UL3	_	-	_
UL4	3.8	44	Near eutrophic
UL5	3.3	42	Near eutrophic
UL6	2.8	41	Near eutrophic
NG1	-	-	_
NG2	18.8	59	-
SK1	0.021	0	-
SK2	< 0.02	0	_

^aTropic state of NG and SK lakes are not stated as number of sampling points are restricted and not spatially distributed

0.50 to 0.55 times lower than the median anion concentrations of MWW samples. The SO_4^{2-} concentrations of IISc groundwater samples and MWW samples are however comparable (10 and 11.5 mg L⁻¹).

Besides K^+ and Na^+ ions, the concentrations of Ca^{2+} , $\mathrm{Mg}^{2+}\text{, Cl}^-\text{, SO}_4{}^{2-}$ and $\mathrm{HCO}_3{}^-$ ions in Bengaluru city groundwater samples are higher than the median MWW values (Table 2). The groundwater in IISc campus is unpolluted as there is no wastewater contamination on the campus. The major ion composition of IISc groundwater samples is considered as representative of non-polluted water source in Bengaluru city. Lithology, weathering of drainage catchment, and rainwater contribute 57 to 98% of Ca²⁺, Mg²⁺, Na⁺, K⁺, HCO₃⁻, SO₄²⁻ and Cl⁻ ion concentrations of unpolluted water source; human activity contributes the remaining 2 to 43% [49]. Geogenic and anthropogenic contributions to the major ion composition of the two sets of groundwater samples are compared in Table 5. The anthropogenic component ratio is calculated as:

Anthropogenic ratio =
$$\frac{Anthro component of Bengaluru city groundwater-Anthro component of IISc groundwater}{Anthro component of IISc groundwater} \times 100\%$$
(2)

The ratios (Table 5) indicate that anthropogenic component of Bengaluru city groundwater samples is 540 to 5333% larger than the corresponding component of IISc groundwater samples. The total annual rainfall of Bengaluru city is 831 mm. The aquifer depths in Bengaluru city range from 3.12 to 21.35 m during monsoon (median value = 7.07 m) and range from 4.8 to 22.01 in the post-pre-monsoon season (median value = 9.8 m) [30]. The main rock formation in the study area is the peninsular gneiss complex, represented by mafic gneiss, biotite gneiss, granodioritic gneiss with intrusions like dykes, and pegmatite. Physical and chemical weathering of the parent rocks form overburden material of varying thickness between the surface and the aquifer. The drainage

characteristics of the overburden is function of soil texture, topography, profile development presence of impervious strata and depth of water table. Based on the above parameters, the drainage of the overburden ranges from excellent (1 to $10^{-3} \text{ cm s}^{-1}$) to poor $(10^{-4} \text{ to } 10^{-6} \text{ cm s}^{-1})$. Owing to the smooth rolling topography of peninsular gneissic terrain. The region exhibits flat (0 to 3%), medium (3–7%) and steep (> 7%) slopes [50]. Occurrence of zones of poor drainage, flat slopes and shallow groundwater makes the Bengaluru aquifers vulnerable to anthropogenic pollution.

Fig. 4 plots the total cation (TZ^+, mN) versus $HCO_3^$ ion concentrations of the lake samples. The data points plot below the 1:1 line imply that the major ion composition of the lake samples is influenced by anthropogenic activities than by the lithology of the drainage catchments [51]. If the major ion composition of Bengaluru lakes was dominated by products of silicate and carbonate weathering, the data points would plot on or close to the 1:1 line [8].

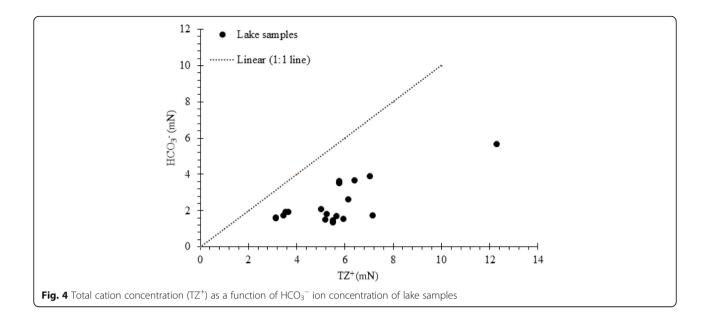
The influence of anthropogenic contamination on the major ion composition of the lakes is examined in the Piper chart (Fig. 5). The diagram shows that the non-polluted IISc groundwater samples dominate in Ca^{2+} and HCO_3^- ions, while the polluted lake samples show dominance of Mg^{2+} , Na^+ , K^+ , Cl^- and SO_4^{2-} ions. Therefore, while, the IISc samples classify as Ca-HCO₃ type, the lake samples classify as mixed Ca-Na-HCO₃ and mixed Ca-Mg-Cl types. Enrichment of Mg^{2+} , Na^+ and Cl^- ions from anthropogenic contamination alters the chemical-type of the lake water. The ionic ratio method (Fig. 4) and Piper chart (Fig. 5) are thus useful tools to understand how factors other than mineral dissolution and hydrochemical facies can impact the chemical composition of water bodies.

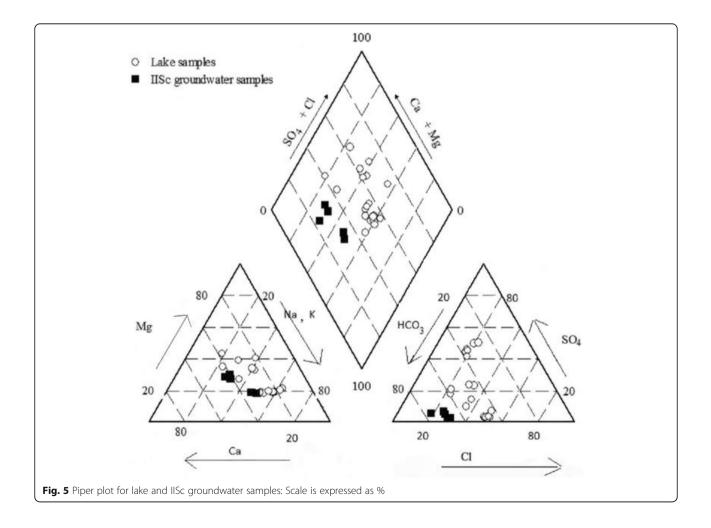
Sodium and chloride ions

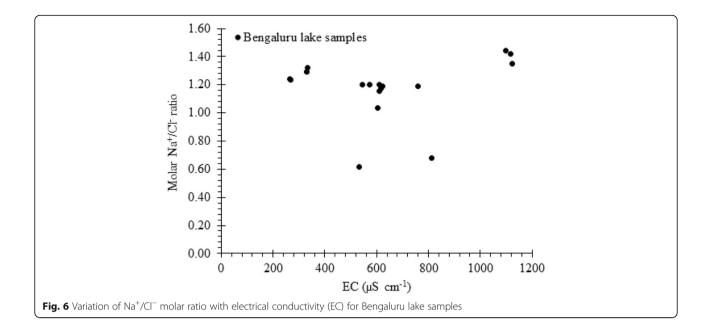
The variation of Na^+/Cl^- molar ratio with EC develops as horizontal scatter (Fig. 6), implying that water

Table 5 Geogenic and anthropogenic contributions to major ion composition of groundwater samples

lon (mg L^{-1})	Bengaluru city gi	roundwater sampl	les [36]	IISc groundwate	Anthropogenic			
	Average value (mg L ^{- 1})	Geogenic component	Anthropogenic component	Average value $(mg L^{-1})$	Geogenic component	Anthropogenic component	components ratio (%) ^a	
HCO3-	341.0	161.7	179.3	165	161.7	3.3	5333	
Cl-	181.0	14.9	166.1	49.7	14.9	34.8	377	
SO_4^{2-}	88.0	5.1	82.9	11.8	5.1	6.7	1133	
Ca ²⁺	117.0	34.6	82.4	38	34.6	3.4	2310	
Mg ²⁺	53.0	13.6	39.4	14.8	13.6	1.2	3226	
Na ⁺	970.0	27.8	69.2	38.6	27.8	10.8	540	
K ⁺	0	-	5.6	_	0	_		



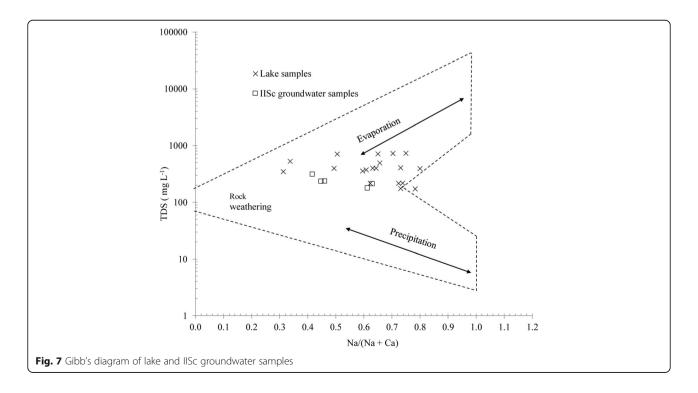


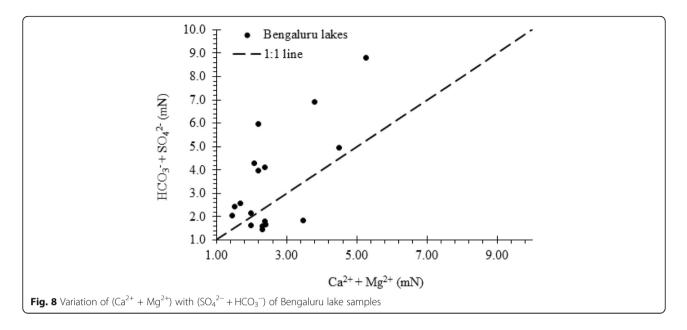


evaporation during dry season strongly impacts the Na-Cl balance in the lakes [8]. Seventy four percent of the lake samples exhibit Na⁺/Cl⁻ molar ratios ranging from 0.62 to 1.29 with median value of 1.2, while 26 % samples exhibit molar ratios ranging from 1.32 to 1.46. Loss of chloride as gaseous HCl is apparently responsible for Na⁺/Cl⁻ ratios being greater than unity. HCl_(gas) is formed from the reaction between sodium chloride (NaCl) and hydrogen ions in the lake water as [52]:

$$NaCl + H^+ \rightarrow Na^+ + HCl_{gas} \tag{3}$$

The Gibb's diagram (Fig. 7) shows that rock weathering dominates the chemical composition of IISc groundwater samples. Comparatively, evaporation process tends to influence the chemical composition of the lake water samples.



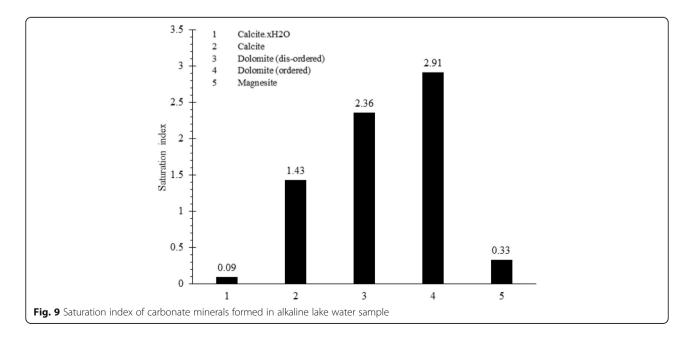


Calcium, magnesium, bicarbonate, sulfate

The $(Ca^{2+} + Mg^{2+})$ vs. $(HCO_3^- + SO_4^{2-})$ chart can reveal the role of mineral dissolution and ion-exchange reactions in controlling the chemical composition of water sources [51, 53]. If the data points are located on the 1:1 line, calcite and gypsum mineral dissolution predominantly contribute to the lake water chemistry. If majority of data points plot in the $(Ca^{2+} + Mg^{2+})$ sector, it is implied that reverse ionexchange enhances the divalent cation concentration in the water. The $(Ca^{2+} + Mg^{2+})$ vs. $(HCO_3^- + SO_4^{2-})$ chart (Fig. 8) of Bengaluru lake samples show that majority of data points are located in the $(HCO_3^- + SO_4^{2-})$ segment, implying a deficit of divalent cations in the lake samples. The saturation index (SI) represents the equilibrium condition between solution and mineral and is calculated as:

$$SI = \log 10 \frac{IAP}{Ksp}$$
 (4)

In Eq. (4), IAP represents the ion activity product of the dissociated chemical species and K_{sp} represents the equilibrium solubility product of the mineral. Calculations of SI of carbonate minerals of a lake water sample at median pH (8.8) showed that the lake is oversaturated (SI > 0) with carbonate mineral (Fig. 9). The



deficiency of divalent cations in the lakes is attributed to their precipitation as carbonate minerals (Fig. 9) and ion-exchange reactions between monovalent ions of sediments and divalent cations in lake water (discussed in section 4.4). Precipitation and ion-exchange reactions increases the relative proportion of Na⁺ to Ca²⁺ ions (median value of ratio = 1.61) in the lake samples.

The Ca²⁺/Mg²⁺ molar ratios of the lake water samples ranged between 0.52 and 0.88 for 37% of the samples and exceeded unity (1.07 to 1.62) for the remaining samples. Dissolution of anthropogenic gypsum [11] apparently contributed to the presence of excess Ca²⁺ ions causing the Ca²⁺/Mg²⁺ molar ratio to exceed unity in many lake samples. In addition, release of SO₄²⁻ ions from gypsum dissolution is possibly responsible for the higher anion concentration (median value = 40 mg L⁻¹) in the lakes than in the MWW samples (median value = 20 mg L⁻¹). About 26,192 gypsum idols were immersed in different lakes of Bengaluru city in 2017 and 2018 [54].

Besides anthropogenic gypsum dissolution, SO_4^{2-} ion concentration in the lakes is contributed by rainwater (3.9 mg L⁻¹, [55]) and MWW contamination. Assuming that the (i) SO_4^{2-} ions in lake water are in equilibrium with ions trapped in the pore solution of the sediments and (ii) dilution effects is balanced by enrichment of the anion concentration from evaporation, it follows that:

SO4 concentration from gyspum dissolution

= (Median SO₄ concentration in lakes)

-(Median SO₄ concentration in MWW

+Mean SO₄ concentration in rainwater) = 40–(20 + 3.9) = 16.1 mg L^{-1}

Mass balance calculations indicate that SO_4^{2-} ions produced from gypsum dissolution (16.1 mg L⁻¹) would release 6.7 mg L⁻¹ of Ca²⁺ ions to the lakes. The median

Table 6 Characteristics of lake sediments

 Ca^{2+} ion concentration (24.5 mg L⁻¹) in the lake samples (Table 2) indicates that gypsum dissolution contributed to 27% of available Ca^{2+} ion concentration in the lake samples; the reminder (73%) is attributed to contributions from MWW contamination and lithology of catchments.

Sink (sediment)-source (lake water) interactions

The PU, UL and NG lake sediments are characterized by organic contents 10.4, 16.2, and 5.8% and gravimetric water contents of 366, 722, and 235% respectively (Table 6). The results indicate that sediments with higher organic matter (OM) are characterized with larger gravimetric water content. The large water holding capacity of humus [45] is apparently responsible for the notable water contents of the sediments (235 to 722%). The large water contents allow dissolution of solutes in the pore solutions of the sediments (TDS ranges from 165 to 301 mg L^{-1}). The pore solutes (determined from water leaching test) of PU and UL sediments (Table 6) are dominated by anions $(Z_{total}^{-} = 122 \text{ to } 129 \text{ mg L}^{-1})$ with major contributions from HCO_3^- and SO_4^{2-} ions. The Ca²⁺ and Mg²⁺ ions mainly contribute to the pore solute cations and the Z⁺_{total} values range from 81 to 88 $mg L^{-1}$. The pore solution of Nagawara sediment has slightly higher concentration of cations $(Z^+_{total} = 3.76)$ meq L^{-1}) than anions (0.79 meq L^{-1}). The sum of anions is less than the sum of cations owing to bicarbonate consumption during algal photosynthesis. The pore solution salinity (TDS) of the sediments (Table 6) is lower than of lake water (Table 2). The resulting chemical concentration gradient favors solute diffusion from lake water into the pore solutions of sediments.

Besides pore solutes, the adsorbed cations of the sediments would also participate in sink-source interactions. The adsorption sites of the sediments are contributed by the exchange sites of mineral fraction and the pH

Lake	Gravimetric	Organic	TDS (from EC test; mg L ⁻¹)	lon compo	lon composition of water leachate (mg L^{-1})							
sediment	water content (%)	matter (%)		Ca ²⁺	Mg ²⁺	Na ⁺	K+	HCO3-	SO4 ²⁻	CI-		
Puttenahalli	366	10.4	286	47.9	33.0	4.2	2.4	73.2	52.5	4.3		
Ulsoor	722	16.2	301	46.3	30.1	2.8	1.9	87.5	34.3	0		
Nagawara	235	5.8	165	4.1	37.2	7.2	5.4	11.2	29.1	0		
				Exchangeable cations (ammonium acetate extract; meq $100 g^{-1}$)								
				Ca ²⁺	Mg ²⁺	Na ⁺	K+					
Puttenahalli				18.8	36	0.52	0.25					
Ulsoor				76.2	51.2	0.7	0.38					
Nagawara				5.1	100.3	0.46	0.66					

(5)

dependent sites of the humus fraction of organic matter [45, 56]. The ammonium acetate extracts of the sediments (Table 6) dominated in Ca²⁺ and Mg²⁺ ions $(Z_{(Ca + Mg) exchangeable} = 55 \text{ to } 127 \text{ meq } 100 \text{ g}^{-1})$. The cation exchange capacity (CEC) of Bengaluru soils range from 1.9 to 22 meq 100 g^{-1} with median value of 12.8 meq 100 g^{-1} [29]. The sum of exchangeable cations (Z⁺_{exchangeable}) of the lake sediments range from 55.6 to 128.5 meq 100 g^{-1} which is > CEC of Bengaluru soils. The results suggest that in addition to the inorganic mineral fraction, the organic coatings on the lake sediments significantly contribute to their exchangeable cation content. The carboxyl, phenolic-OH, amino and carbonyl groups of the humic and non-humic substances in lake sediments develop pH dependent charges and significantly contribute to the cation adsorption sites [45, 56]. Ion-exchange reactions between adsorbed Na⁺ and K⁺ ions of lake sediments with Ca²⁺ and Mg²⁺ ions of lake water possibly enriched the adsorbed divalent cation content of sediment particles as [57]:

$$2NaX + Ca^{2+} \rightarrow 2Na^{+} + CaX_2 \tag{6}$$

The ion-exchange reaction (Eq. 6) would also contribute to depletion of divalent cation concentration in the lakes.

Implications of MWW contamination on lake water classification

Table 7 provides an excerpt of water quality parameters used to classify water resources [58] for water supply with conventional treatment (Class IIA), or for recreational use with body contact (Class IIB). In the present study, Malaysian water quality index [58] is chosen over Indian surface water quality standards to enable inclusion of COD parameter (indicator of organic contamination) for the lake classification. The Indian surface water quality standards do not specify tolerance limits for COD [59]. The median values of the Bengaluru lakes indicate that owing to high COD (> 25 mg L⁻¹), high pH (> 8.7) and DO (> 5–7 mg L⁻¹), the lakes do not categorize as class IIA or IIB. The presence of nonpermissible Al^{3+} ion concentrations (> 0.2 mg L⁻¹) renders the lake water toxic. Presence of sewage in the MWW can be expected to impart fecal coliform counts $>400 \text{ CFU } 100 \text{ mL}^{-1}$ in the lakes. Besides, alterations in major ion chemistry, organic and microbial contamination, biochemical reactions associated with photosynthesis activity of algae and release of toxic metals from mineral dissolution are the consequences of contamination of Bengaluru lakes. Stopping the flow of partially treated or untreated sewage in the lakes is essential to restore their physical, chemical and biological characteristics.

Conclusions

Anthropogenic activity impacted the major ion composition of Bengaluru lakes and aquifers. The concentrations of most major ions in Bengaluru city groundwater are higher than the corresponding MWW values. Occurrence of zones of poor drainage, flat slopes and shallow groundwater makes the Bengaluru aquifers vulnerable to anthropogenic pollution. The ionic ratio plot of Bengaluru lakes implies that the major ion composition of the lake samples is influenced by anthropogenic activities than by the lithology of the drainage catchments indicating that outfall of sewage treatment plant and outlets of stormwater drains have contaminated the Bengaluru lakes.

Enrichment of Mg^{2+} , Na^+ and Cl^- ions from anthropogenic contamination changed the hydrochemical-facies of the lake water. The lake samples classify as mixed Ca-Na-HCO₃ and mixed Ca-Mg-Cl types, while, nonpolluted IISc groundwater samples classify as Ca-HCO₃ type of water. Rock-weathering influences the chemical composition of non-polluted IISc groundwater samples, while, evaporation tends to dominate the chemical characteristics of lake water. Interactions of NaCl particles with atmosphere during evaporation altered the Na/Cl balance of the lake samples. The alkaline pH due to photosynthetic metabolism of algae facilitated precipitation of carbonate minerals and a deficit of Ca and Mg

 Table 7 Classification of lakes based on select parameters [58]

Parameter	Class II A: Water supply with conventional treatment	Class IIB: Recreational use with body contact	Median concentration of Bengaluru Lakes
Ammoniacal Nitrogen (mg L^{-1})	0.3	0.3	0
COD (mg L^{-1})	25	25	41
DO (mg L^{-1})	5–7	5–7	12.5
рН	6.5 to 9.0	6.5–9.0	9.1
Electrical Conductivity (μ S cm ⁻¹)	1000	-	610
Faecal Coliform (CFU 100 mL^{-1})	100	400	Not tested

ions in the lakes. Ion-exchange reactions between adsorbed cations of lake sediments and lake water solutes also contributed to deficit of divalent cations in the lakes. Dissolution of anthropogenic gypsum contributed to 27% of available Ca^{2+} ion concentration in the lake samples and the reminder (73%) is contributed by MWW contamination and lithology of drainage catchments. Besides alterations in major ion chemistry, organic contamination, biochemical reactions associated with photosynthesis activity of algae and release of toxic Al^{3+} ions from chemical weathering of lake sediments are the consequences of lake contamination.

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Authors' contributions

Sudhakar Rao (SR), Priscilla A (PA), Nitish VM (NVM), Lydia A (LA) designed the field sampling and field/laboratory measurements. PA, NVM and LA performed the field sampling and experiments. SR, PA, NVM and LA were involved in analysis and interpretation of the results. SR wrote the manuscript in consultation with PA, NVM and LA. The author(s) read and approved the final manuscript.

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Availability of data and materials

All data used in the study appear in the submitted article.

Competing interests

The authors declare that they have no competing interests.

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